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## Vibrational study of nitrogen incorporation in InGaAsN alloys

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**Abstract.** We present an infra-red (IR) reflectivity and Raman study of  $\text{In}_x\text{Ga}_{1-x}\text{As}_{1-y}\text{N}_y$  ( $x = 0$  and  $\sim 0.08$ ,  $y \sim 0.03$ ) alloys grown by molecular beam epitaxy. We observed: — strong diagonal Raman components of GaAs-type phonons, indicating a local trigonal distortion of the alloy lattice induced by nitrogen ordering; — indium induced splitting of the Ga-N type vibration, indicating a formation of different local nitrogen atomic arrangements; — a sharp (halfwidth  $\sim 1 \text{ cm}^{-1}$ ) IR mode at  $360 \text{ cm}^{-1}$ , of unknown origin.

### Introduction

The InGaAsN alloys have recently attracted considerable attention as promising materials for laser diodes in the  $1.3\text{--}1.5 \mu\text{m}$  range as well as more efficient solar cells. These applications exploit the unusual electronic property — a “giant bowing”, which arises from the large electronegativity and very small ion radii of the nitrogen [1, 2], together with the possibility of perfect lattice matching to GaAs substrates. Up to now little is known about the micro/nanostructure, i.e. possible atomic ordering and phase separation, of these alloys, which can dramatically alter their electronic and transport properties important for device applications. In our previous paper [3] using Raman spectra we revealed local trigonal distortion of coherently strained  $\text{GaAs}_{1-x}\text{N}_x$  ( $x \sim 0.02$ ) alloys, indicating the existence of [111] GaAs/GaN ordering. In present paper using Raman and IR vibrational spectroscopy we study of the properties of N incorporation into InGaAsN alloys lattice matched to GaAs.

### 1. Experiment

Coherently strained  $\text{GaAs}_{1-y}\text{N}_y$  and lattice matched  $\text{In}_x\text{Ga}_{1-x}\text{As}_{1-y}\text{N}_y$  ( $x \sim 0.08$ ,  $y \sim 0.03$ ) layers with thickness  $\sim 0.2 \mu\text{m}$  were grown by solid source MBE on (001) semi-insulating GaAs substrates at temperature  $450^\circ\text{C}$ . An RF-plasma source was used to generate atomic nitrogen from  $\text{N}_2$ . The composition was determined by calibration growths of GaAsN and InGaAs along with x-ray diffraction. The layers have strong photoluminescence at room temperature at  $1.05\text{--}1.15 \text{ eV}$ .

We also used MOCVD grown reference samples of  $\text{In}_x\text{Ga}_{1-x}\text{As}$  ( $x \sim 0.1$ ) and  $\text{GaAs}_{1-y}\text{N}_y$  ( $y \sim 0.03$ ) studied by us previously in [4].

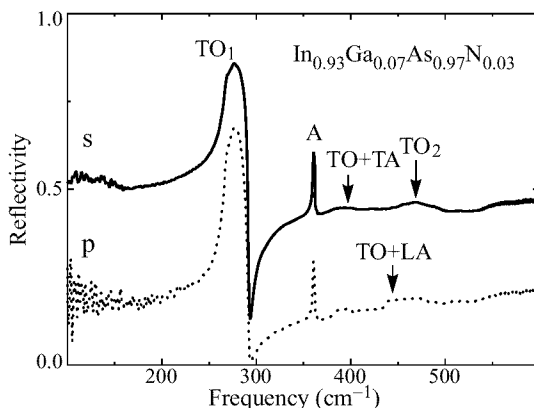
Polarized room temperature IR spectra were taken at oblique incidence ( $45^\circ$ ) with a Bruker IFS-66V spectrometer.

The Raman spectra (RS) were measured in the backscattering geometry using a double monochromator DFS-52 and excited with a  $488 \text{ nm}$   $\text{Ar}^+$ -ion laser at room temperature. The  $xy$  and  $xx$  (where the  $x \parallel [100]$  and  $y \parallel [010]$ ) components of RS excited from the (001) growth plane were analyzed. According to Raman selection rules of zinc blende structure

only optical phonons with longitudinal (LO) polarization are allowed in  $xy$  configuration, while all optical modes are forbidden in the  $xx$  one.

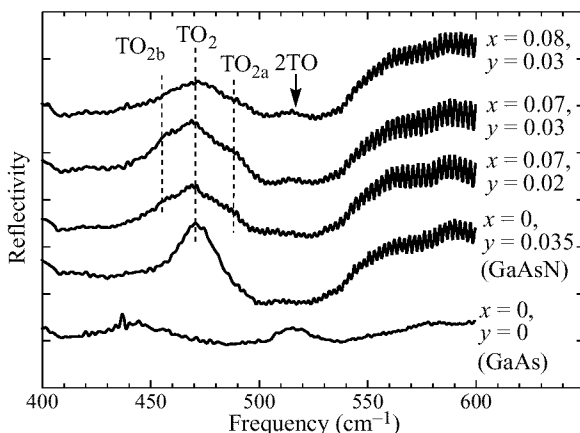
## 2. Results and discussion

The results of our IR and Raman measurements are presented in Figs. 1–4. As can be seen from Fig. 1, the IR spectra of  $\text{In}_{0.07}\text{Ga}_{0.93}\text{As}_{0.97}\text{N}_{0.03}$  alloys have a main intense restrahl band  $270\text{--}290\text{ cm}^{-1}$  due to the GaAs-type phonons, a very sharp peak A at  $360\text{ cm}^{-1}$ , and several weak bands in the range  $400\text{--}550\text{ cm}^{-1}$ , corresponding to second order spectra of GaAs-type. The Ga-N type phonon  $\text{TO}_2$  also observed at  $470\text{ cm}^{-1}$ .

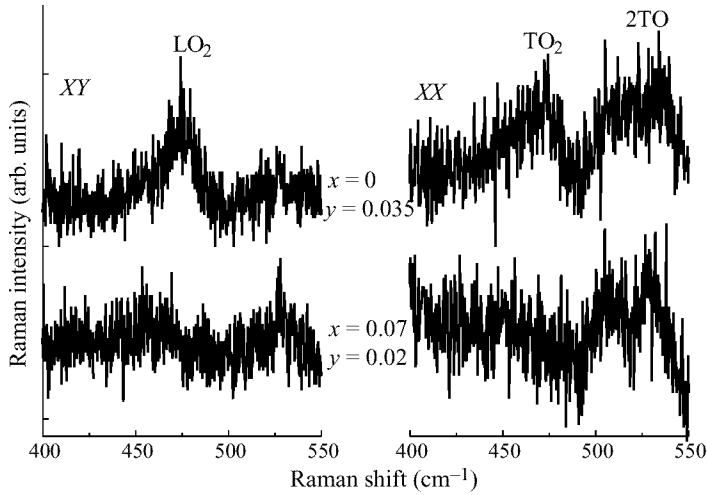


**Fig. 1.** Polarized (s — solid, p — dashed curves) IR reflection spectra of  $\text{In}_{0.07}\text{Ga}_{0.93}\text{As}_{0.97}\text{N}_{0.03}$  alloys.

The A peak has unusually small halfwidth  $1.5\text{ cm}^{-1}$ , which is much smaller than the typical phonon damping parameter of binary III–V compounds ( $\sim 3\text{ cm}^{-1}$ ). We observed this IR peak in all GaAsN and InGaAsN layers grown by MBE, but it was absent in the IR spectra of the MOCVD grown GaAsN layers. On the other hand, our Raman measurements did not reveal any band at  $360\text{ cm}^{-1}$ , what makes the origin of this peak at the moment unclear.



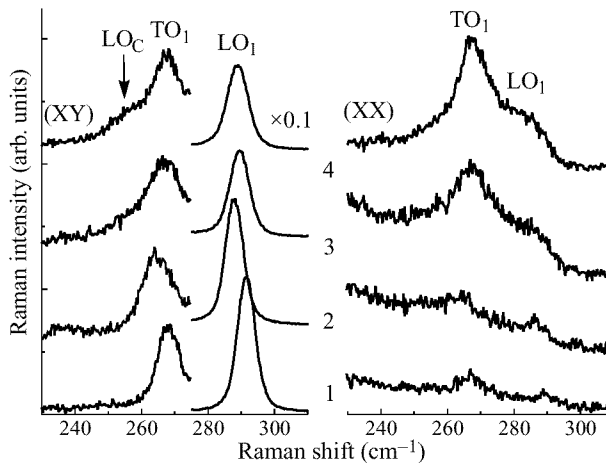
**Fig. 2.** s-polarized IR reflection spectra of  $\text{GaAs}_{1-y}\text{N}_y$ ,  $\text{In}_x\text{Ga}_{1-x}\text{As}_{1-y}\text{N}_y$  alloys and GaAs substrate in  $400\text{--}600\text{ cm}^{-1}$  frequency range.



**Fig. 3.**  $xy$  (left) and  $xx$  (right) Raman components of Ga-N type mode of  $\text{In}_{0.07}\text{Ga}_{0.93}\text{As}_{0.02}\text{N}_{0.98}$  and  $\text{GaAs}_{0.965}\text{N}_{0.035}$ .

In the IR spectra we observed the GaN-type vibration ( $\text{TO}_2$ ) in both  $\text{InGaAsN}$  and  $\text{GaAsN}$ , as can be seen from Fig. 2, where we compare the spectra of three  $\text{In}_x\text{Ga}_{1-x}\text{As}_{1-y}\text{N}_y$  ( $x \sim 0.08$ ,  $y \sim 0.03$ ) and one  $\text{GaAs}_{1-y}\text{N}_y$  ( $y \sim 0.035$ ) layers with the spectra of  $\text{GaAs}$ . It can be seen that in  $\text{InGaAsN}$  the  $\text{TO}_2$  band is much broader. It reveals a fine structure consisting of a low —  $\text{TO}_{2b}$  and a high —  $\text{TO}_{2a}$  frequency shoulder split by  $\sim 15 \text{ cm}^{-1}$  from the central  $\text{TO}_2$  peak. The splitting clearly indicates that there are three different local atomic arrangements of nitrogen in the lattice of  $\text{InGaAsN}$ , which is induced by the presence of indium.

In the Raman spectra the Ga-N type mode was observed only in  $\text{GaAsN}$  (see Fig. 3). As in our previous study [3] it appears in both zinc blende allowed —  $xy$  and forbidden —  $xx$  configurations, which is due to a strong local trigonal distortion of the  $\text{GaAsN}$  lattice



**Fig. 4.**  $xy$  (left) and  $xx$  (right) Raman components of GaAs-type modes of  $\text{GaAs}$  (1),  $\text{In}_{0.14}\text{Ga}_{0.86}\text{As}$  (2),  $\text{In}_{0.07}\text{Ga}_{0.93}\text{As}_{0.02}\text{N}_{0.98}$  (3) and  $\text{GaAs}_{0.965}\text{N}_{0.035}$  (4) alloys.

caused by the bond relaxation and ordering of GaN<sub>3</sub>As microclusters ( [111] GaN/GaAs ordering).

While we did not detect the GaN-type mode in InGaAsN we did get evidence that the GaN<sub>3</sub>As microcluster ordering is preserved in the presence of In. This is seen in Fig. 4, where the strong enhancement (compared with GaAs and InGaAs) of the forbidden  $xx$  scattering of the GaAs-type TO<sub>1</sub> phonon is observed for both GaAsN and InGaAsN.

The appearance of the strong zinc blende forbidden Raman components, indicating [111] GaN/GaAs ordering, is an anomalous property of InGaAsN, which was not observed in conventional III–V alloys. The results presented show that it does not depend on the macroscopic strain due to lattice mismatch between epi layer and substrate. The driving force for the ordering can be strong local strains (bond relaxation) caused by the small nitrogen radii.

#### *Acknowledgments*

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